

Absorption and emission of polariton modes in a ZnSe-ZnSSe heterostructure

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We investigate the absorption and emission of a 25 nm ZnSe layer, which was grown on a GaAs buffer and cladded by ZnSSe layers. Due to the coupling of light with the exciton resonances polariton modes propagate through the ZnSe layer. Their interferences appear as additional peaks in the reflection spectra and can be explained by the effect of spatial dispersion. We present additional experimental results for the emission of the sample after excitation by a pump pulse, showing corresponding interference peaks of the polariton modes, whose maxima strongly decrease to higher energies. Our exact theoretical analysis shows that the ratio of emission and absorption is given by the population of the globally defined states of the electromagnetic field, i.e. the polariton distribution which is generated by the pump pulse. This distribution, being far from thermal quasi-equilibrium, shows pronounced peaks at the polariton modes depending on the energy of the pump pulse.

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INTRODUCTION

In the vicinity of the band edge strong coupling of light with exciton resonances leads to a splitting of the light dispersion, known as polariton effect. Besides the well-known Fabry-Perot modes the spatial dispersion leads to an additional series of interferences of polariton waves above the exciton resonances [1]. The behavior of amplitude and phase of the reflected light around these resonances was investigated in [2] for a 25 nm ZnSe layer, which was grown on a GaAs buffer and cladded by ZnSSe layers. Characteristic changes of the phase were found around the polariton interferences, which in particular are sensitive to the damping of the excitonic resonances. The theoretical description was based on a broadened Elliot formula [3] for the susceptibility of the heavy-hole (hh) and light-hole (lh) exciton resonances in the active ZnSe layer including the spatial dispersion. Pekar's additional boundary conditions (ABC's) [4] were applied and according to the microscopic approach in [5] a weak penetration of the polarization into the cladding layers was taken into account. The good agreement of the experimental results for the reflection with our theoretical model enables to reconstruct the absorbed intensity of a probe pulse in the sample, while the transmitted beam is completely absorbed in the GaAs buffer.

In this paper we additionally investigate the light emission of the sample at low excitation. The experimental results are presented in the following section. A quantum mechanical and many-body description of emission was given in [7, 8] based on Keldysh's technique for photon Green's functions (GF's). However, in this approach spatial dispersion was neglected. A theoretical description of the emission including spatial dispersion is a non-trivial problem. In [9, 10] the problem of energy conservation was discussed in the context of the dielectric approximation (DA). Recently, starting from energy conservation, one of us (K.H.) [11] presented an exact proof that the ratio between incoherent emission e and coherent absorption a is given by the polariton distribution b which for quasi-equilibrium tends towards a Bose distribution. In section we give a short outline of this approach and demonstrate that the theoretical concept agrees with the experimental findings.

EXPERIMENTAL RESULTS

Our experiments were performed at a sample grown by molecular beam epitaxy and composed of a 25 nm ZnSe layer, cladded by two 1 μm ZnS_xSe_{1-x} layers with a sulfur content of $x = 7\%$. The sample was contained in a He-flow cryostat and cooled to a temperature of $T = 10$ K. The experiments were carried out using 120 fs (FWHM) pulses with a spectral linewidth of 15 meV (FWHM) centered at different photon energies in order to cover the whole spectral domain between the heavy- and light-hole exciton resonances of the ZnSe layer. For details of the measurement of the reflection we refer to [2]. The emission was detected after excitation with pump pulses, which were focused on the surface of the sample with a spot diameter of 50 μm under perpendicular incidence. The emitted light was measured under an angle of 4° by a triple additive grating spectrometer with a spectral resolution of 0.1 meV. The emission

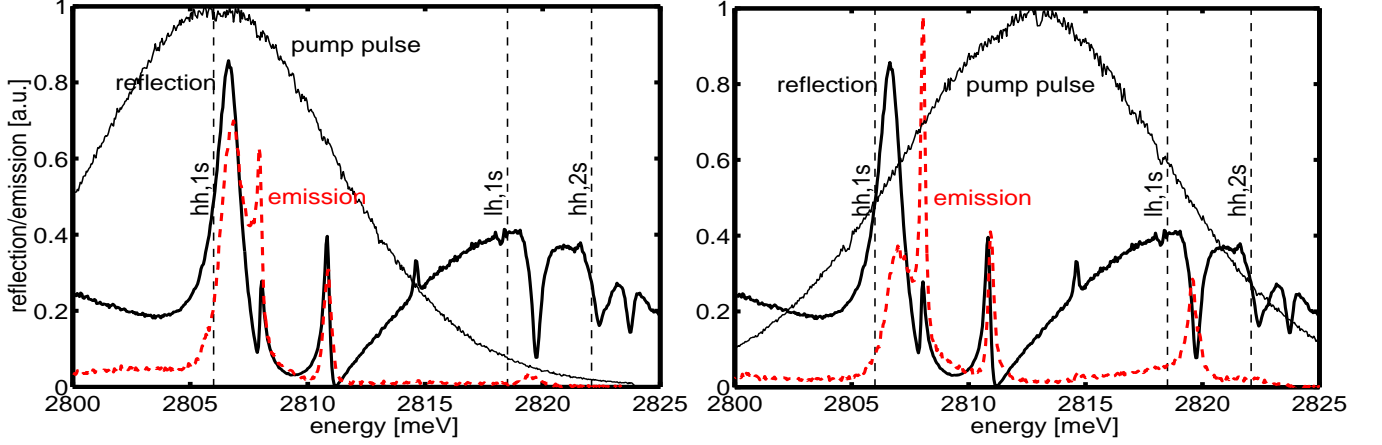


FIG. 1: Reflection (solid black line) and emission (dashed red) for two different pump pulses (thin black).

was recorded by integrating the spectrometer's CCD over a train of many pump pulses. The pulse repetition rate is 80MHz, which corresponds to a repetition period of 13 ns.

The results of our measurements are presented in Fig. 1. The spectrum of reflectivity is given by solid black lines in both parts of the figure. The emitted intensity after pumping with pulses (thin black lines) centered at two different energies (left: at the hh-exciton, right: between hh- and lh-exciton) is presented with dashed red lines. The emission is scaled to be comparable with the reflectivity. Additionally, the positions of the exciton resonances are marked by vertical dashed lines. Generally, the emission is concentrated on the polariton interference modes. However, the amplitude of the emission peaks strongly decreases with increasing energy. In [1] it was found that in dependence of the geometry of the sample even or odd polariton modes appear more or less pronounced. As one can see from the figure, this may be different concerning reflectivity and emission. While in the reflection the first peak directly above the 1s-hh exciton is much higher than the second one, in the emission the weight is shifted more to the second peak. We demonstrated in our theoretical analysis in section that the shape of the emission is related to the distribution function of excited polariton states, which are generated by the pump pulse.

THEORETICAL DESCRIPTION

General balance between absorption and emission

In this section we start with a general description of wave propagation through a slab of finite thickness L with the aim to demonstrate that there is a general connection between emission and absorption, summarizing those points of the approach in [14] which are important for the understanding of the present paper. The slab is considered to be infinitely extended in the transverse y - z -direction, and homogeneously excited. For notational simplicity, TE-polarized light propagating freely in the transverse direction is considered. Due to cylindrical symmetry around the x -axis, the transverse vector potential $\mathbf{A}(\mathbf{r}, t)$ can be chosen in the z -direction. Assuming steady state conditions, the propagation equation for the averaged field, after Fourier transforming with respect to $(y, z) \rightarrow \mathbf{q}_\perp$ and $t - t' \rightarrow \omega$, in this geometry has the structure

$$\int dx' D^{\text{ret}, -1}(x, x') A(x') = 0 \quad , \quad (1)$$

where $D^{\text{ret}, -1}$ is the inverse of the retarded photon GF

$$D^{\text{ret}, -1}(x, x') = \left(\frac{\partial^2}{\partial x^2} + q_0^2 \right) \delta(x - x') - P^{\text{ret}}(x, x'). \quad (2)$$

Here and in what follows, the variables \mathbf{q}_\perp and ω , which enter all equations parametrically only, are omitted where possible. In vacuum the propagation in x -direction is governed by $q_0^2 = [(\omega + i\delta)/c]^2 - q_\perp^2$. The retarded polarization function of the medium $P^{\text{ret}}(x, x', q_\perp, \omega)$, acting in Eq. 2 as selfenergy of photons, is related to the susceptibility χ

according to

$$P^{\text{ret}}(x, x') = -\frac{\omega^2}{c^2} \chi(x, x'). \quad (3)$$

The forward propagating solution of the homogeneous equation (1) has the structure

$$A(x) = \begin{cases} e^{iq_0 x} + r e^{-iq_0 x} & \text{for } x < -\frac{L}{2} \\ t e^{iq_0 x} & \text{for } x > \frac{L}{2}, \end{cases} \quad (4)$$

where r, t are reflection and transmission coefficients for the field amplitudes. Due to the symmetry $x \leftrightarrow -x$, $A(-x)$ is a solution, too (backward propagating, i.e., incidence from right). It is the advantage of this approach that it works in general, without specifying the solutions inside the slab. Of course, these solutions have to be determined to calculate the coefficients t , and r . This will be done in section . Following Poynting's theorem, the x -component of the Poynting vector in the considered geometry is related to the absorbed field energy $W = j \cdot E$ by

$$S\left(\frac{L}{2}, \omega\right) - S\left(-\frac{L}{2}, \omega\right) = - \int_{-L/2}^{L/2} dx W(x, \omega). \quad (5)$$

The current density is related via $j = \partial P / \partial t$ to the polarization $P = \chi E$, and the electric field via $E = -\partial A / \partial t$ to the vector potential A . Assuming a monochromatic wave of frequency ω_0 incident in the $(q_0, \mathbf{q}_{\perp,0})$ -direction

$$A(x, \mathbf{q}_{\perp}, \omega) = \frac{1}{2} [A_0(x, \mathbf{q}_{\perp,0}, \omega_0) \delta(\omega - \omega_0) \delta_{\mathbf{q}_{\perp}, \mathbf{q}_{\perp,0}} + A_0^*(x, \mathbf{q}_{\perp,0}, \omega_0) \delta(\omega + \omega_0) \delta_{\mathbf{q}_{\perp}, -\mathbf{q}_{\perp,0}}], \quad (6)$$

in Eq. (5) after straightforward calculation for each $\omega_0 \rightarrow \omega$ and $\mathbf{q}_{\perp,0} \rightarrow \mathbf{q}_{\perp}$ yields

$$1 - |r|^2 - |t|^2 = a = \frac{i}{2q_0} \hat{P}, \quad (7)$$

$$\hat{P} = \int dx dx' A^*(x) \hat{P}(x, x') A(x'). \quad (8)$$

Eq. (7) simply balances the incoming intensity (~ 1) into absorption a and the sum of the reflected and transmitted intensity. On the other hand, due to the well-known GF identities [13]

$$\hat{P} = P^{\text{ret}} - P^{\text{adv}} = P^> - P^<, \quad (9)$$

the absorption balances generation $iP^>(x, x')$ and recombination $iP^<(x, x')$ of excitons in the medium.

The incoherent or correlated emission is defined as the one without external sources, i.e. for vanishing averaged fields. In this case, due to the non-commuting field operators, the symmetrized Poynting vector $\mathbf{S} = \frac{1}{2\mu_0} (\mathbf{E} \times \mathbf{B} - \mathbf{B} \times \mathbf{E})$ and absorption/emission $W = \frac{1}{2} (\mathbf{j} \cdot \mathbf{E} + \mathbf{E} \cdot \mathbf{j})$, respectively, have to be used. Following the approach in Ref. [7] the intensity of the emitted light is given by

$$S(x) = \frac{1}{2} \left\{ \frac{\partial}{\partial x'} [D^>(x, x') + D^<(x, x')] \right\}_{x'=x}, \quad (10)$$

and the emitted field energy by

$$W(x) = \int dx' [P^>(x, x') D^<(x', x) - P^<(x, x') D^>(x', x)]. \quad (11)$$

$D^>(x, x', \mathbf{q}_{\perp}, \omega)$ and $P^>(x, x', \mathbf{q}_{\perp}, \omega)$ are the Keldysh components of the photon GF and of the polarization function (see Eq. (9)), respectively. Here as in Eqn's (1-9) the variables \mathbf{q}_{\perp} and ω are introduced by Fourier transform, but are dropped, since they only appear as parameters.

The polarization function

$$P^>(x, x') = P_m^>(x, x') \mp i\delta \frac{2\omega}{c^2} \Theta(\pm\omega) \delta(x - x') \quad (12)$$

comprises a medium part P_m and a part which describes the vacuum as an infinitely weak ($\delta \rightarrow 0$) absorber [8]. As shown in [14], the formal solution (optical theorem) of the photon Dyson equation can be used in (11) yielding after straightforward calculation the incoherent absorption as

$$\mathcal{W} = \int dx W(x) = -\frac{i}{q_0} \mathcal{P}^<, \quad (13)$$

As usual, a distribution function b will be attributed by definition to the global recombination $\mathcal{P}^<(\omega, \mathbf{q}_\perp)$ defined in (13)

$$\mathcal{P}^< = \int dx dx' A^*(x) P_m^<(x, x') A(x') = b \hat{\mathcal{P}}. \quad (14)$$

Now Poynting's theorem (5) provides the incoherent energy flow $e = S(L/2) = -S(-L/2)$ from the incoherent absorption (13): $2e = -\mathcal{W}$. Then from (8), (13), and (14), for any frequency $\omega > 0$ and propagation direction $\mathbf{q} = (q_0, \mathbf{q}_\perp)$, we obtain for the spectrally and directionally resolved energy flow (emission)

$$e = \frac{i}{2q_0} \mathcal{P}^< = b a. \quad (15)$$

The distribution b is accessible to direct observation in experiments measuring the (incoherent) emission and the classical absorption, i.e., transmission and reflection. It generalizes Planck's formula for the black body radiation to the non-equilibrium radiation of an excited medium in the steady state. Relation (15) is generally valid. It was derived without specifying the fields inside the slab. In this respect it works as a criterion which has to be fulfilled for each model applied to the polariton pulse propagation, in particular for all applied ABC's in the calculations. In contrast to the authors in [9, 10], we find energy conservation valid in the DA, too [14].

For quasi-equilibrium, due to the Kubo-Martin-Schwinger condition [15], the distribution $b(\omega, \mathbf{q}_\perp)$ develops into a Bose distribution $b(\omega) = (\exp[\beta(\hbar\omega - \mu)] - 1)^{-1}$ with the temperature T ($\beta = 1/kT$) and the chemical potential μ , being independent of \mathbf{q}_\perp . Measuring emission and absorption enables to check whether quasi-equilibrium is realized.

Absorption and Emission of the investigated sample

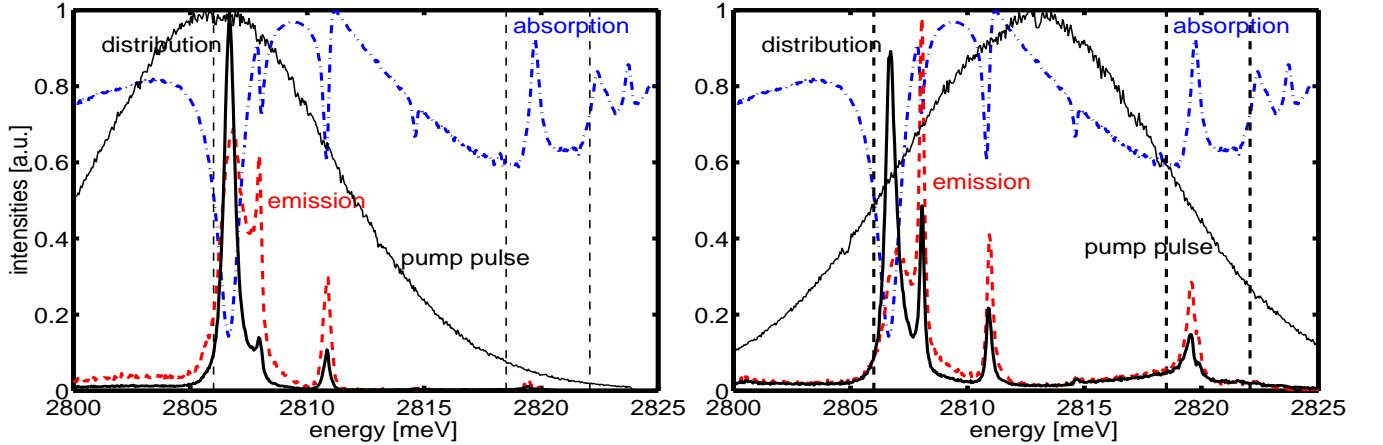


FIG. 2: Absorption (dash-dotted blue line), emission (dashed red) and distribution function (solid black) for two different pump pulses (thin black).

In our sample absorption and emission is investigated in the vicinity of the heavy-hole (hh) and light-hole (lh) exciton resonances of the 25 nm ZnSe layer which is cladded by two 1 μm ZnSSe layers. As we have demonstrated in [2], the reflection is dominated by polariton interferences (see Fig. 1, 2) and can be described considering a dielectric function for the active ZnSe-layer, which contains the 1s-hh, 2s-hh, and 1s-lh exciton resonances including their spatial dispersion. The polariton dispersion splits into four branches and additional boundary conditions (ABC's) for the single polariton waves at the surfaces have to be considered. The experiments fit well applying Pekar's ABC's and

assuming a small increased effective ZnSe layer (25.5 nm) which takes into account the penetration of the excitonic wave function into the cladding layers. This treatment is a simple way to circumvent the costly numerical microscopic calculation of the spatially resolved polarization presented in [5] and the approach in [6]. The experimental results could not be verified applying Ting's ABC's [16]. However, the dielectric approximation [9, 10], being a combination of differently weighted ABC's of Ting and Pekar, gives good agreement, since the weight is strongly focused to Pekar's ABC's.

The good agreement of our theoretical calculations with the experimental results in Fig. 1 (see also [2]) encourages us to calculate the absorbed intensity of light according to Eq. (7). In contrast to the single slab considered in the preceding section no light passes through the back side of the sample, since it is completely absorbed in the GaAs buffer layer. That is to say, there is no total transmission, and the absorption in Eq. (7) is simply $a = 1 - R$, where R is the intensity of the reflected light normalized to the intensity of the incoming light. The total absorption of the sample is presented in Fig. 2 by the dash-dotted blue lines. The pump pulses and the emission correspond to those in Fig. 1. Additionally, the quotient of emission and absorption is given (solid black line), which represents the distribution function b of polariton states (15) in the sample. It shows clearly that the polariton modes are predominantly occupied. Depending of the position of the pump pulse, the occupation of the single polariton modes varies. This behavior indicates that the detected emission comes rather from the excited polariton states, generated by the pump pulse, than from a quasi-equilibrium distribution.

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